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Development of MMC Gamma Detectors for Nuclear Analysis

C. R. Bates, C. Pies, S. Kempf, L. Gastaldo, A. Fleischmann, C. Enss, S. Friedrich

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C.R. Bates · C. Pies · S. Kempf · L.
Gastaldo · A. Fleischmann · C. Enss · S.
Friedrich

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Abstract Non-destructive assay (NDA) of nuclear materials would benefit from gamma detectors with improved energy resolution in cases where line overlap in current Ge detectors limits NDA accuracy. We are developing metallic magnetic calorimeter (MMC) gamma-detectors for this purpose by electroplating 150 μm thick Au absorbers into microfabricated molds on top of Au:Er sensors. Initial tests under non-optimized conditions show an energy resolution of 200 eV FWHM at 60 keV. Monte Carlo simulations illustrate that this resolution is starting to be sufficient for direct detection of ^{242}Pu in plutonium separated from spent nuclear fuel.

Keywords MMC · gamma ray

1 Introduction

One of the primary tools used in nuclear safeguards to verify the operating history of a nuclear reactor is to compare the operators declaration to spent fuel characteristics. This is typically done using ratios of high intensity gamma ray lines with a high purity germanium (HPGe) detector. This method is becoming less reliable as advances in materials engineering make it possible to get more energy out of a fuel assembly before failure. As the fuel assembly is irradiated for longer periods of time the correlation of the high intensity gamma ray lines to fuel characteristics weakens. In addition, high burn-up produces increasing amounts of ^{242}Pu , which can currently only be measured indirectly³. If the ratios of all Pu isotopes could be determined more accurately than what is possible with HPGe detectors,

C.R. Bates
Lawrence Livermore National Laboratory
7000 East Ave. L-188 Livermore, CA 94550, USA
Tel.: +1-925-424-6763
E-mail: bates26@llnl.gov

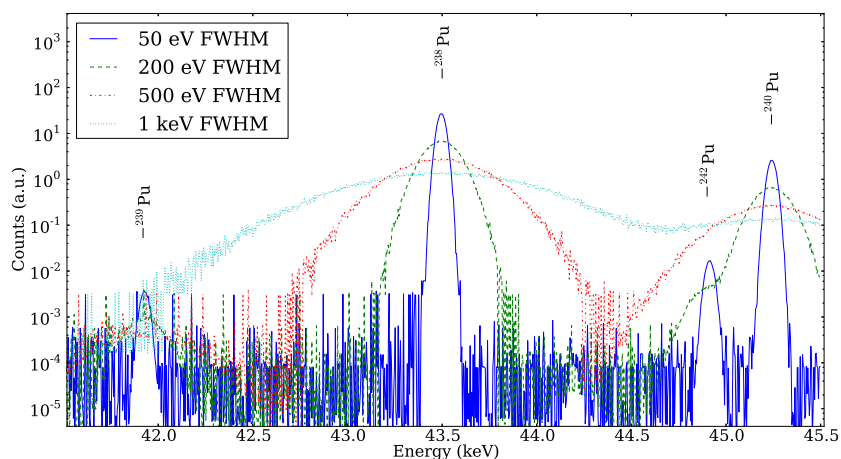


Fig. 1 Geant4 Simulation of Pu gamma ray lines near 40 keV for different detector resolution's, assuming a Pu point source and full separation of Pu from the fission and activation products of the spent fuel. (color figure online)

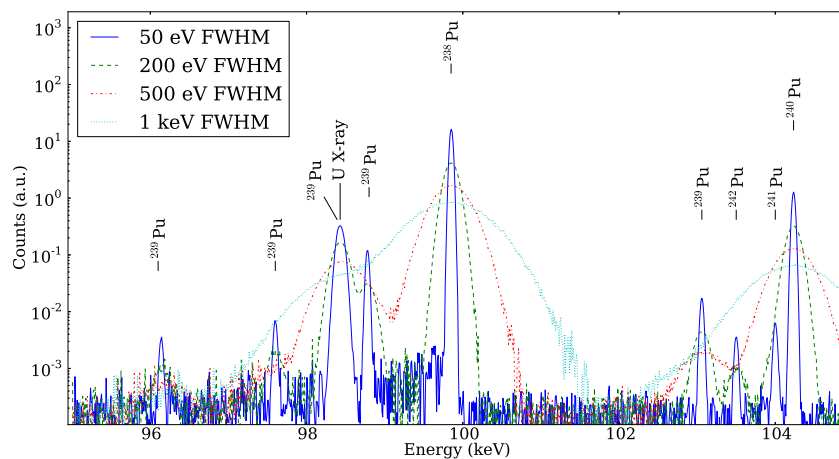


Fig. 2 Geant4 Simulation of Pu gamma ray lines near 100 keV for different detector resolution's, assuming a Pu point source and full separation of Pu from the fission and activation products of the spent fuel. (color figure online)

better constraints could be placed on initial fuel enrichment and reactor operating history. This makes the development of high-resolution MMC gamma detectors desirable for use in this application.

2 Direct Measurement of ^{242}Pu in spent fuel

In order to measure the different Pu isotopes it is currently necessary to chemically separate the Pu from other metals. This is already common for routine mass

spectroscopy and is also a standard part of fuel reprocessing. The separated Pu is typically deposited onto a thin Al planchet to minimize the scattering in the source and placed in front of the detector outside of the cryostat.

We have performed Geant4 Monte Carlo simulations to determine the resolution requirements of our detector⁵. As an input source we used the program ORIGEN-ARP to generate the expected Pu content for a fuel assembly with a typical irradiation profile for a modern pressurized water reactor. In order to simplify the simulation was assumed a point source geometry and that the Pu was perfectly separated from other elements. We focused on the regions around 40 keV and 100 keV, since they contain gamma rays from all the relevant Pu isotopes and the proximity of the lines enables ratio measurements with a simple correction for efficiency.

Figure 1 shows the simulated spectrum from this area of interest with several different detector resolutions. The 1 keV FWHM spectra is typical of a commercial coaxial HPGe detector while the 500 eV FWHM is typical of a thin planar HPGe optimized for low energy spectroscopy. The 200 eV FWHM spectrum shows the limit at which the ^{242}Pu line at 44.8keV can be resolved, which is comparable to the best MMC spectrum in this work. Finally the 50 eV FWHM is comparable to what is theoretically achievable in an optimally designed MMC that would have appreciable efficiency at 40 keV. Based on statistics, we estimate that a resolution of 75 eV FWHM is required to determine the Pu-242 content to 1% precision, because the interfering Pu-240 line is roughly 100 times stronger and separated by only 300 eV⁶. This precision depends on the Pu-242 / Pu-240 ratio and the Compton background level, which may be higher than in Figure 1 because of the simplifying assumptions made in the simulations.

Figure 2 shows the simulated spectra for the same four detectors in the 100 keV region, which is widely used for Pu analysis⁴. Again, an MMC energy resolution of 200 eV starts to separate the individual Pu emissions, although it may be more difficult to measure ^{242}Pu in this region even with 50 eV FWHM. This is because the line is an order of magnitude weaker and the secondary Pu $\text{K}_{\alpha 1}$ X-ray fluorescence at 103.75 keV can provide strong interference in concentrated Pu samples.

3 Gamma-ray measurements of ^{241}Am

In order to demonstrate the feasibility of the Pu measurement with an MMC we installed an ^{241}Am source in front of a Heidelberg maXs-200 MMC in a 18 mK dilution fridge operated at the Kirchhoff Institute for Physics⁷. This design uses a Au:Er sensor deposited on top of a 0.5 mm x 2 mm meander of Nb, which is used to both apply the field and readout the signal. In order to be sensitive to hard X-rays there is a 150 μm thick electroplated Au absorber on top of the Au:Er. This device was read out using a two-stage SQUID, model C6XS116 from Magnicon GmbH. The pulses were saved using a 12-bit Gage data acquisition card. The raw pulses were then processed using an optimal filter routine. Despite wiring issues with the device that forced us to operate with approximately 10% of the designed magnetizing current it was still possible to achieve a resolution of 200eV at 60 keV (Figure 4). This performance shows that it should be possible to directly measure the ^{242}Pu line using this device even in sub-optimal operating

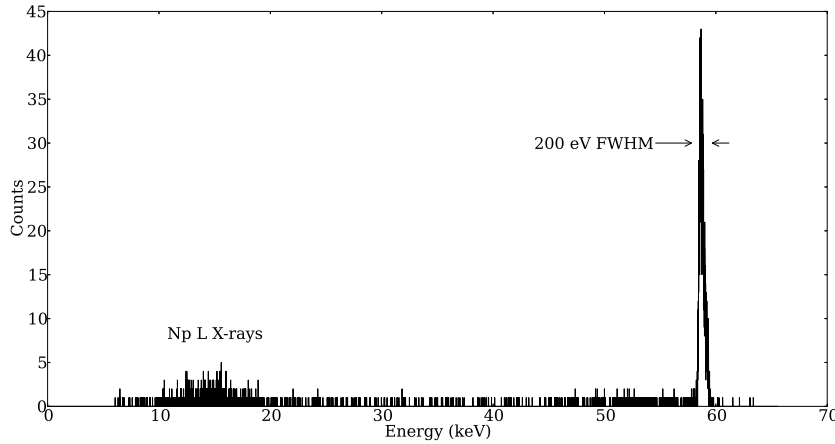


Fig. 3 Spectra of ^{241}Am taken with a maXs-200 MMC.

conditions. Under optimal conditions this device has been shown to achieve an energy resolution of 60 eV FWHM at 60 keV⁷. We are currently working on attaining similar resolution.

4 Outlook

In order to perform high accuracy plutonium analysis arrays of MMCs will be required to achieve good statistics. This is due to the long decay time, and therefore limited countrate of the devices, on the order of milliseconds and several hundred hertz respectively. In order for larger arrays to be feasible it is necessary for the individual pixels to have high yield. This is currently limited by the difficulty in removing the mold for the absorbers, made out of SU-8 photoresist. We are developing a process based on AZ-125 nXT photoresist which we have successfully spin coated to a thickness of 300 μm . This photoresist has been shown to produce thick molds for electroplating without the problems associated with SU-8⁸. This should simplify the fabrication of high-yield MMC arrays with thick electroplated absorbers in the future.

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